

Amendments to the Specification

Please replace the paragraph at page 2, lines 20 through 26 with the following amended paragraph:

In a specific embodiment, the coating on the core has an empirical formula of $\text{Li}_{x1}\text{A}_{x2}\text{Ni}_{1-y1-z1}\text{Co}_{y1}\text{Q}_{z1}\text{O}_a$. “x1” is greater than about 0.1 and equal to or less than about 1.3. “x2,” “y1” and “z1” each is greater than 0.0 and equal to or less than about 0.2. “a” is greater than about 1.5 and less than about 2.1. “A” is at least one element selected from the group consisting of barium, magnesium, calcium and strontium. [“B”] “Q” is at least one element selected from the group consisting of boron, aluminum, gallium, manganese, titanium, vanadium and zirconium.

Please replace the paragraph at page 5, line 27 through page 6, line 7 with the following amended paragraph:

In one embodiment, the coating has the empirical formula of: $\text{Li}_{x1}\text{A}_{x2}\text{Ni}_{1-y1-z1}\text{Co}_{y1}\text{Q}_{z1}\text{O}_a$. The crystal structure of the core, and optionally of the coating, is of an “ α -NaFeO₂-type” and can be characterized as having a hexagonal lattice within the R-3m group. The composition of the invention is suitable for employment as the active material in cathodes for rechargeable lithium-ion or lithium polymer-type batteries, among other uses. The invention also generally is directed to lithium batteries and to cathodes of lithium batteries that employ the composition of the invention, and to methods of forming the composition.

Please replace the paragraph at page 7, lines 4 through 8 with the following amended paragraph:

In still another embodiment, the core material has an empirical formula, $\text{Li}_x\text{Mg}_y\text{Ni}_{1-y}\text{Co}_y\text{B}_v\text{O}_2$ $\text{Li}_{x1}\text{A}_{x2}\text{Ni}_{1-y1-z1}\text{Co}_{y1}\text{Q}_{z1}\text{O}_a$. “x” is greater than about 0.1 and equal to or less than about 1.3. “y” is greater than 0.0 and equal to or less than about 0.4. “v” is equal to or less than about 0.4. [“B”] “Q” is at least one member of the group consisting of manganese, boron, aluminum and gallium.

Please replace the paragraph at page 7, lines 11 through 19 with the following amended paragraph:

In still another embodiment of the invention, the coating has an empirical formula $\text{Li}_{x1}\text{Mg}_{x2}\text{Ni}_{y1}\text{Co}_{y2}\text{B}_z\text{O}_a$. “x1” is greater than about 0.1 and equal to or less than about 1.3. “x2,” “y1” and “z1” each is greater than about 0.0 and equal to or less than about 0.2. “a” is greater than about 1.5 and less than about 2.1. “A” is at least one element selected from the group consisting of barium, magnesium, calcium and strontium. [[“B”]] “Q” is at least one element selected from the group consisting of boron, aluminum, gallium, manganese, titanium, vanadium and zirconium. In one preferred embodiment, “A” is magnesium and [[“B”]] “Q” is aluminum. In another preferred embodiment, “A” is magnesium and [[“B”]] “Q” is manganese.

Please replace the paragraph at page 7, line 20 through page 8, line 2 with the following amended paragraph:

In one embodiment, the invention is a composition, such as a crystal, wherein a core and a coating of the composition each has an empirical formula: $\text{Li}_{x1}\text{Mg}_{x2}\text{Ni}_{y1}\text{Co}_{y2}\text{B}_z\text{O}_a$, wherein “x1” is greater than about 0.1 and equal to or less than about 1.3, “x2,” “y1” and “z1” each is greater than about 0.0 and equal to or less than about 0.2, and “a” is greater than about 1.5 and less than about 2.1. “A” is at least one element selected from the group consisting of barium, magnesium, calcium and strontium, and [[“B”]] “Q” is at least one element selected from the group consisting of aluminum, gallium, manganese, titanium, vanadium and zirconium. The coating has a greater ratio of cobalt to nickel than the core. In one preferred embodiment, “A” is magnesium and [[“B”]] “Q” is aluminum. In another preferred embodiment, “A” is magnesium and [[“B”]] “Q” is manganese. In still another embodiment, “A” is magnesium and [[“B”]] “Q” is gallium.

Please replace the paragraph at page 10, line 18 through page 11, line 5 with the following amended paragraph:

Although not wishing to be bound by any particular theory, the crystal structure of crystals of the invention are believed to be of “ α - NaFeO_2 -type,” and is in the following described in a hexagonal lattice within a “R-3m” space group. In this structure, Li and “A” atoms occupy the so-called “3a” site in the structure ($x=0$, $y=0$, and $z=0$), the Ni and [“B”] “Q” atoms occupy the “3b” site ($x=0$, $y=0$, $z=0.5$), and oxygen occupies the “6c” site ($x=0$, $y=0$, $z=z$). The “z” coordinate of oxygen changes depending on the nature of “A” and [“B”] “Q” elements and their relative ratios. Typically the “z” coordinate is between 0.22 and 0.26. The cell parameters of this material, “a” and “c,” also varies with the chemical composition. Typically, cell parameters are found between the following ranges: $a=2.75 - 2.95 \text{ \AA}$, and $c=13.9 - 14.3 \text{ \AA}$. Generally, the cell parameters exhibit a gradient through the particle structure. Also there is a gradient of diminishing nickel concentration from the core to the surface of each particle. The gradient particulate structure typically produces a diffraction pattern that has asymmetric Bragg reflection peaks when subjected to X-ray diffraction (XRD). This peak asymmetry is a “fingerprint” of the gradient material of the invention and provides experimental evidence that the material has a gradually changing elemental composition.

Please replace the paragraph at page 11, lines 6 through 19 with the following amended paragraph:

Coatings of the cathode materials of the current invention can improve the safety of Ni-based materials at the same time as capacity is maintained or improved, although the Ni-atoms are diluted. This is made possible by the simultaneous substitution of atoms in both the Li-site (3a-site) and the Ni-site (3b site). “A” atoms partly substitute for Li atoms in the 3a site [“B”] “Q” atoms substitute for Ni in the 3b site. Further, the capacity and cyclability is higher due to higher stabilization of the structure than that found in the LiCoO_2 system. This means that more lithium can be removed from the structure without having a structural collapse. It is believed that safety is high compared to LiNiO_2 due to shielding of the 3b-3b interaction, which is the dominating mode for structural collapse. This enables less structure damage upon cycling and higher stability at the lower Li contents at about $x=0.2$, which is the traditional cut-off for the LiNiO_2 systems, especially for compounds containing about 20% Co. Polarization behavior of the composition of the invention can lead to higher capacity for devices that utilize a 3.5 volt cut-off for the electronics.

Please replace the paragraph at page 12, line 17 through page 13, line 2 with the following amended paragraph:

The structural reason for this collapse is described below. The crystal structure of layered LiMO_2 is shown in Figure 2. Intuitively, it is not obvious that the crystal of Figure 2 will collapse, i.e. drastically reduce the distance between the MO_2 layers, when Li ions are removed from the structure. One would expect that the negatively charged neighbors (oxygen atoms) repel after the positively charged ions are removed. This should cause quite an opposite effect; the crystal should expand. However, this is shown not to be the case. Instead it is instructive to look at in the (012) family of crystallographic planes in the LiMO_2 structure, which are shown in Figure 2. The sequence of these planes is depicted in Figure 3. The crystal consists of alternating slightly distorted hexagonal metal/oxygen layers. The metal layer consists of both Li and ~~M (Co/Ni/B)~~ M (Co/Ni/Q) atoms, while the oxygen layer contains only oxygen atoms. This view of the structure helps to understand why the crystal is not repelling when the Li-ion layers are removed even in a simplistic ionic model. In fact, as seen from Figure 3, when the Li atoms are removed from the structure, M-atoms still remain in the metal plane and keep the structure stable.

Please replace the paragraph at page 13, lines 2 through 10 with the following amended paragraph:

The crystal is kept together by interaction of oxygen and metal planes. The metal planes consist of alternating rows of Li/A and ~~Ni/Co/B atoms~~ Ni/Co/Q atoms. When Li atoms are removed, the metal plane becomes partially depopulated, which should result in an overall decrease in structural stability. Another conclusion that we have drawn simply from structural considerations is that, when Li is removed, it leaves behind a quasi 1-dimensional string of ~~M(Ni,Co,B)~~ M(Ni,Co,Q) atoms in the (012) plane which should be thermodynamically less stable and thus should have a strong tendency to diffuse into sites that were formerly occupied by Li atoms.

Please replace the paragraph at page 14, lines 3 through 22 with the following amended paragraph:

The change in the c-axis length upon Li deintercalation has been modeled. The results for Li_xCoO_2 are presented in Figure. 7. A drastic change in the slope of the $c(x)$ function takes place at around $x=0.5$, which coincides with the maximum degree of Li deintercalation from LiCoO_2 . It is believed that this change shows the limit for stability in the crystal structure and determines the maximum capacity of a cathode material. In other words, the slope of the $c(x)$ function in the low- x region is an indicator of relative capacity of a material, i.e. when this slope is larger, the “critical point” in x moves towards lower x values, indicating greater stability of a material. The c-axis change for LiNiO_2 also was modeled, and is shown in Figure. 8. The change in the slope of $c(x)$ for LiNiO_2 takes place at much lower values of x , indicating increased capacity due to the increased structural stability of LiNiO_2 as compared to LiCoO_2 . The underlying physics behind this behavior is believed to be based on the fact that Ni has one electron more than Co. This additional electron occupies so-called antibonding states, which counteract the tendency to collapse the structure by Ni-Ni bonds, shown in Figure. 5. This feature is used as a rationale for doping with ~~B atoms~~ Q atoms. It is believed that doping of ~~B atoms~~ Q atoms into Ni sites lowers the number of electrons that are occupying antibonding orbitals, thereby improving safety due to the dilution of Ni atoms. However, due to the compromised capacity, “A” atoms are used to further stabilize the structure, thereby allowing relatively high capacity while maintaining safety. Further, due to the increased structural stability, cyclability typically is high.

Please replace the paragraph at page 14, line 23 through page 15, line 7 with the following amended paragraph:

During discharge, polarization of an active cathode material is especially important. For instance, in cellular telephones, traditionally electronics has stopped working at voltages below 3.5V. For this application it is important that as much capacity as possible is available above this limit. Figure 9 shows a polarization comparison between LiCoO_2 and $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ oxide materials. As can be seen from Figure. 9, the total capacity of the $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ material is higher if the cell is allowed to discharge below 3.5V. However, if 3.5V is the cut-off voltage during discharge, LiCoO_2 would have a relatively better capacity. This feature also is addressed in the current invention. We have found certain dopants to increase voltage during discharge. Elements that will positively influence the voltage profile when doped on the 3b site (~~B atoms~~ Q atoms) are among the preferred elements. Element that increase polarization compared to pure

LiNiO_2 oxide when substituted on the 3b site are manganese, boron, aluminum, gallium and cobalt. Manganese exhibits a particularly good effect on safety properties, while high capacity can be maintained for low substitution levels.